Dispersive forces on bodies and atoms: a unified approach

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A unified approach to the calculation of dispersive forces on ground-state bodies and atoms is given. It is based on the ground-state Lorentz force density acting on the charge and current densities attributed to the polarization and magnetization in linearly, locally, and causally responding media. The theory is applied to dielectric macro- and micro-objects, including single atoms. Existing formulas valid for weakly polarizable matter are generalized to allow also for strongly polarizable matter. In particular when micro-objects can be regarded as single atoms, well-known formulas for the Casimir-Polder force on atoms and the van der Waals interaction between atoms are recovered. It is shown that the force acting on medium atoms—in contrast to isolated atoms—is in general screened by the other medium atoms.

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I. INTRODUCTION

It is well known that polarizable particles and macroscopic bodies—matter whose electromagnetic properties are described terms of macroscopic quantities—are subject to forces in the presence of electromagnetic fields, even if the (fluctuating) fields vanish on average and the bodies do not carry any excess charges and are unpolarized. In particular, this is also the case when the field-matter system can be assumed to be in its ground state, where only quantum fluctuations are responsible for the forces. In this case it is common to speak of vacuum forces or dispersive forces, which obviously represent a genuine quantum effect. Basically, it can be distinguished between three kinds of dispersive forces, namely van der Waals (vdW), Casimir-Polder (CP), and Casimir forces, depending on whether forces between individual particles, between particles and macroscopic bodies or between macroscopic bodies are respectively considered (see, e.g., Ref. [1] for an overview).

Since both CP forces on atoms and Casimir forces on macroscopic bodies may be regarded as being macroscopic manifestations of microscopic van der Waals forces, intimate relations between them can be expected. Nevertheless, quite different theoretical approaches to the two kinds of forces have been developed. Compared to the large body of work in this field, only little attention has been paid to their common origin and consequential relations between them (see, e.g., Refs. [2, 3, 4, 5, 6]). Moreover, the studies have been based on specific geometries such as simple planar structures, and weakly polarizable matter has been considered.

More attention has been paid to the relations between Casimir forces and van der Waals forces, but again for specific geometries and weakly polarizable matter (see, e.g., Refs. [2, 7, 8, 9, 10, 11, 12]). Quite recently, a very general relation between CP forces (as calculated within the frame of macroscopic QED) and multi-atom van der Waals forces has been established [13], where it has been shown that the CP force acting on an atom in the presence of a dielectric body of given permittivity can be regarded as being the sum of all many-atom van der Waals forces with respect to the atoms of the body.

In this paper we develop, within the framework of QED in linearly, locally, and causally responding magnetodielectric media, a unified approach to the calculation of dispersive forces acting on ground-state macro- and micro-objects. Since the origin of any electromagnetic force is the Lorentz force acting on appropriate charges and currents, we first consider the ground-state expectation value of the Lorentz force density acting on the charge and current densities attributed to the polarization and magnetization fields of linear media, taking fully into account the noise polarization and noise magnetization that are associated with absorption. From the ground-state Lorentz force density obtained in this way, the force acting on an arbitrary body or an arbitrary part of it can be then obtained by integration over the respective volume. Applying the theory to dielectric systems, we present a very general force formula, the applicability of which ranges from dielectric macro-objects to micro-objects, also including single atoms, without restriction to weakly dielectric material. In particular, this formula enables us to extend the well-known CP-type formula for the force acting on a weakly dielectric (micro-)object to an arbitrary one.

The paper is organized as follows. In Sec. II the theory of CP forces acting on single ground-state atoms in the presence of magnetodielectric bodies is recapitulated. The ground-state expectation value of the Lorentz force density in linear magnetodielectric media is calculated in Sec. III. On this basis, in Sec. IV the force acting on an arbitrary dielectric body or a part of it is derived. The theory is applied in Sec. V to the calculation of the force acting on a dielectric micro-object, the limiting case of a single atom (which can be either an isolated atom or a medium atom) is considered, and contact to earlier results found for planar structures is made. In Sec. VI it is shown that the theory can also be used to study the van der Waals interaction between two atoms. Finally, a summary and some concluding remarks are given in Sec. VII. For the sake of clarity, some of the derivations

are given in appendices.

II. CASIMIR-POLDER FORCE

The CP force acting on a ground-state atom in the vicinity of arbitrary magnetodielectric bodies that linearly, locally, and causally respond to the electromagnetic field can be regarded as being a conservative force. Hence it can be given by the negative gradient of a potential which in the leading order of perturbation theory reads (see, e.g., [14, 15, 16, 17, 18])

$$\mathbf{F}^{(\mathrm{at})}(\mathbf{r}) = -\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi), \quad (1)$$

where $\mathbf{r}$ is the position of the atom, $\alpha(i\xi)$ is its polarizability, and $\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi)$ is the scattering part of the classical retarded Green tensor $\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)$ taken at imaginary frequencies. Note that the scattering part of the Green tensor contains all the necessary information about the configuration of the magnetodielectric bodies, whose electromagnetic properties are characterized by the electric and magnetic susceptibilities that are complex functions of frequency and may vary in space. The (translationally invariant) bulk part of the Green tensor, which would diverge in the coincidence limit, is not needed in Eq. (1), because it cannot contribute to the force. Equation (1), which can be derived [18] on the basis of exact quantization of the macroscopic electromagnetic field in linearly, locally, and causally responding media [19], strictly applies to isolated atoms, but not to medium atoms nor to guest atoms in a substrate medium. Note that the atomic ground-state polarizability $\alpha(\omega)$ in leading-order perturbation theory,

$$\alpha(\omega) \sim \sum_k \frac{\Omega_k^2}{\omega_k^2-\omega^2}\,, \quad (2)$$

features poles on the real frequency axis due to the neglect of level broadening. If necessary, the correct response function properties [20] may be restored by means of an appropriate limit prescription, viz.

$$\alpha(\omega) \sim \lim_{\gamma\to 0+}\sum_k \frac{\Omega_k^2}{\omega_k^2-\omega^2-i\gamma\omega}\,. \quad (3)$$

In order to apply Eq. (1) to a dielectric body of volume V_M, let us consider instead of a single atom a collection of atoms that are (strictly) contained inside a space region of volume V_M, and let us add up the individual forces as given by Eq. (1). Since the mutual interaction of the atoms is completely ignored in this way, it is clear that this method gives only the lowest-order approximation to the total force. If the number density of the atoms (defined on a suitably chosen macroscopic length scale) is denoted by $\eta(\mathbf{r})$, the total force in this approximation reads

$$\mathbf{F} = -\frac{\hbar\mu_0}{2\pi}\int_{V_\mathrm{M}}\mathrm{d}^3r\int_0^\infty \mathrm{d}\xi\,\xi^2\eta(\mathbf{r})\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi). \quad (4)$$

Since the validity of Eq. (4) obviously requires sufficiently weakly polarizable atoms and/or a sufficiently low number density of atoms, the collection of atoms can be viewed as dielectric matter of volume V_M and small susceptibility

$$\chi_\mathrm{M}(\mathbf{r},i\xi) = \eta(\mathbf{r})\alpha(i\xi)/\varepsilon_0, \quad (5)$$

which implies that the permittivity of the overall system has slightly been changed by $\delta\varepsilon(\mathbf{r},i\xi)=\chi_\mathrm{M}(\mathbf{r},i\xi)$. In particular, applying Eq. (4) to a dielectric micro-object whose number density of atoms is constant over the small volume V_M, we obtain the force

$$\mathbf{F} = -V_\mathrm{M}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi). \quad (6)$$

Clearly, application of Eq. (4) to dielectric bodies (including micro-objects) that are dense and/or consist of strongly polarizable atoms becomes questionable. Hence, the problem of a generalization of Eq. (4) to arbitrary dielectric bodies or parts of them arises. As we will see, a satisfactory answer can be given on the basis of the ground-state Lorentz force density in media.

III. GROUND-STATE LORENTZ FORCE

As shown in Ref. [21], the Casimir force between (linearly, locally and causally responding) macroscopic bodies can be regarded as the expectation value of the Lorentz force acting on the charges and currents attributed to the polarization and magnetization of the bodies. To be more specific, the charge and current densities $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}(\mathbf{r})$, respectively, which the Lorentz force density acts on are given by

$$\hat{\rho}(\mathbf{r}) = \int_0^\infty \mathrm{d}\omega\,\underline{\hat{\rho}}(\mathbf{r},\omega) + \mathrm{H.\,c.} \quad (7)$$

and $\hat{\mathbf{j}}(\mathbf{r})$ accordingly, with

$$\underline{\hat{\rho}}(\mathbf{r},\omega) = -\varepsilon_0\boldsymbol{\nabla}\cdot\{[\varepsilon(\mathbf{r},\omega)-1]\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\}+(i\omega)^{-1}\boldsymbol{\nabla}\cdot\underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r},\omega) \quad (8)$$

and $[\kappa_0=\mu_0^{-1}]$

$$\begin{aligned}\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) &= -i\omega\varepsilon_0[\varepsilon(\mathbf{r},\omega)-1]\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\\ &\quad+\boldsymbol{\nabla}\times\{\kappa_0[1-\kappa(\mathbf{r},\omega)]\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega)\}+\underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r},\omega). \end{aligned} \quad (9)$$

Here, $\underline{\hat{\mathbf{j}}}_\mathrm{N}(\mathbf{r},\omega)$ is the (fluctuating) noise current density that acts as a Langevin noise source in the (macroscopic) Maxwell equations in the ω−domain, $\varepsilon(\mathbf{r},\omega)$ and $\kappa^{-1}(\mathbf{r},\omega)$ are respectively the (complex) permittivity and

permeability, and $\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega)$ are the (positive) frequency components of the medium-assisted electromagnetic field:

$$\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) = i\mu_0\omega \int \mathrm{d}^3r'\, \mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{10}$$

$$\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = \mu_0 \boldsymbol{\nabla}\times \int \mathrm{d}^3r'\, \mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega) \tag{11}$$

(for details of the quantization scheme, see Ref. [19]). The expectation value of the Lorentz force,

$$\mathbf{F} = \int_{V_{\mathrm{M}}} \mathrm{d}^3r \int_0^\infty \mathrm{d}\omega \int_0^\infty \mathrm{d}\omega' \,\langle \underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega') + \underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\times \underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle_{\mathbf{r}'\to\mathbf{r}}, \tag{12}$$

taken with respect to the ground state of the linearly interacting field-matter system then yields the Casimir force acting in the zero-temperature limit on the material in a chosen spatial region of volume V_{M}. Again, divergent bulk contributions must be discarded in the limit $\mathbf{r}'\to\mathbf{r}$, as they would correspond to an unphysical "self-forces" of the respective volume elements.

The Casimir force as given by Eq. (12) [together with Eqs. (8)–(11)] can be equivalently rewritten as a surface integral over a stress tensor,

$$\mathbf{F} = \int_{\partial V_{\mathrm{M}}} \mathrm{d}\mathbf{a}\cdot \mathsf{T}(\mathbf{r}), \tag{13}$$

where (at zero temperature)

$$\mathsf{T}(\mathbf{r}) = \lim_{\mathbf{r}'\to\mathbf{r}} \left[\mathsf{S}(\mathbf{r},\mathbf{r}') - \tfrac{1}{2}\mathsf{1}\mathrm{Tr}\,\mathsf{S}(\mathbf{r},\mathbf{r}')\right] \tag{14}$$

together with

$$\mathsf{S}(\mathbf{r},\mathbf{r}') = -\frac{\hbar}{\pi}\int_0^\infty \mathrm{d}\xi\, \left[\frac{\xi^2}{c^2}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi) + \boldsymbol{\nabla}\times \mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi)\times \overleftarrow{\boldsymbol{\nabla}}'\right]. \tag{15}$$

Here and in the following, $\mathsf{1}$ denotes the unit tensor and $\times\overleftarrow{\boldsymbol{\nabla}}'$ is meant to act to its left. With regard to the controversial view held in Ref. [22], it should be emphasized that Eq. (13) together with Eqs. (14) and (15) yields the genuine electromagnetic Casimir force acting on bodies or pieces of them. Clearly, in mechanical equilibrium the Casimir force is balanced by additional internal or external (mechanical) forces that are not included in the equations considered here.

Let us return to Eq. (12) together with Eqs. (8)–(11). It is not difficult to show (Appendix A) that Eqs. (8) and (9) can be rewritten as

$$\underline{\hat{\rho}}(\mathbf{r},\omega) = \frac{i\omega}{c^2}\,\boldsymbol{\nabla}\cdot\int \mathrm{d}^3r'\, \mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{16}$$

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = \left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times - \frac{\omega^2}{c^2}\right)\int \mathrm{d}^3r'\, \mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r}',\omega), \tag{17}$$

and that the relation

$$\langle \underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}_{\mathrm{N}}^\dagger(\mathbf{r}',\omega')\rangle = \frac{\hbar}{\mu_0\pi}\,\delta(\omega-\omega')\left\{\frac{\omega^2}{c^2}\,\mathrm{Im}\,\varepsilon(\mathbf{r},\omega)\mathsf{1}\delta(\mathbf{r}-\mathbf{r}') - \boldsymbol{\nabla}\times[\mathrm{Im}\,\kappa(\mathbf{r},\omega)\mathsf{1}\delta(\mathbf{r}-\mathbf{r}')]\times\overleftarrow{\boldsymbol{\nabla}}'\right\} \tag{18}$$

holds for the ground-state expectation value. Employing standard properties of the Green tensor (see, e.g., Ref. [19]), from Eqs. (10), (11) and (16)–(18) it follows that

$$\langle \underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle = \frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\boldsymbol{\nabla}\cdot\mathrm{Im}\,\mathsf{G}(\mathbf{r},\mathbf{r}',\omega) \tag{19}$$

and

$$\langle \underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle = -\frac{\hbar}{\pi}\,\delta(\omega-\omega')\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathrm{Im}\,\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\times\overleftarrow{\boldsymbol{\nabla}}', \tag{20}$$

which can be used to express the integrand of the volume integral in Eq. (12)—that is, the Casimir force density—in terms of the (scattering part of the) Green tensor solely. Extension of the theory to include thermal states is straightforward.

IV. FORCE ON DIELECTRIC BODIES

To illustrate the theory, we first apply Eq. (12) [together with Eqs. (19) and (20)] to the calculation of the Casimir force acting on a dielectric body in some space region of volume V. We want to study—mostly in parallel—the two cases sketched in Figs. 1 and 2, namely (i) an isolated body (Fig. 1), and (ii) a body that is an inner part of some larger body (Fig. 2). In both cases, arbitrary magnetodielectric bodies are allowed to be present in the outer region V_{B} in the figures.

Let $\varepsilon(\mathbf{r},\omega)$ be the permittivity of the system in the absence of the dielectric matter in V and assume that it changes to $\varepsilon(\mathbf{r},\omega)+\Delta\varepsilon(\mathbf{r},\omega)$ when the additional dielectric matter is introduced into the initially empty space region V. Here and in the following, ΔA denotes the exact change of a quantity $A[\varepsilon(\mathbf{r},\omega)]$ due to a given (not necessarily small) change of the permittivity, $\Delta\varepsilon(\mathbf{r},\omega)$, whereas the notation δA is used to indicate the familiar first-order variation of A produced by a small variation $\delta\varepsilon(\mathbf{r},\omega)$. Further, let $\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)$ and $\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)$ be the Green tensors of the system in the cases where the dielectric matter inside the space region V is present and absent, respectively, with both of them taking into account the magnetodielectric bodies in the space region V_{B} in Figs. 1 and 2. We may then write

$$\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega) = \mathsf{G}(\mathbf{r},\mathbf{r}',\omega) + \Delta\mathsf{G}(\mathbf{r},\mathbf{r}',\omega), \tag{21}$$

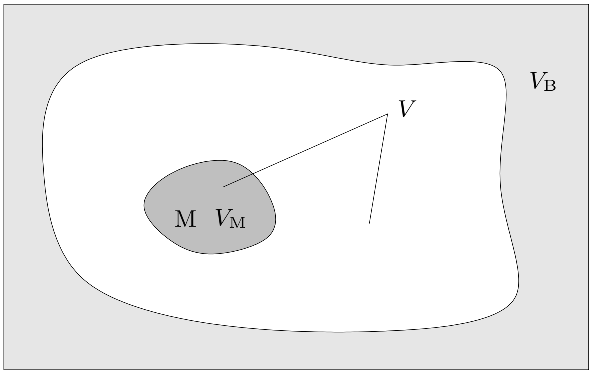

FIG. 1: A dielectric body M of volume V_{M} inside an empty-space region of total volume V. There may be arbitrary magnetodielectric bodies in the outer region V_{B}.

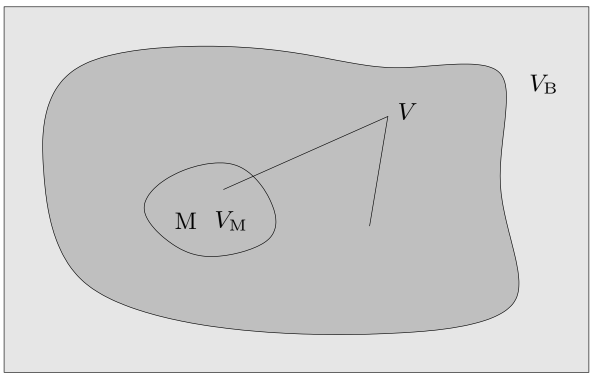

FIG. 2: A dielectric body M of volume V_{M} that is an inner part of a larger dielectric body of volume V. There may be arbitrary magnetodielectric bodies in the outer region V_{B}.

where the change of the Green tensor, $\Delta\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)$, due to the introduction of the dielectric matter into V obeys the Dyson-type equation

$$\Delta\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)=\frac{\omega^2}{c^2}\int \mathrm{d}^3s\,\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\Delta\varepsilon(\mathbf{s},\omega)\mathsf{G}_V(\mathbf{s},\mathbf{r}',\omega), \tag{22}$$

with the integral running over the space region V (at most).

To calculate the changes of the expectation values $\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle$ [Eq. (19)] and $\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle$ [Eq. (20)] $(\mathbf{r}\in V)$, we note that for $\mathbf{r}\in \bar{V}$ the Green tensor $\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)$ satisfies the same differential equation as the free-space Green tensor, i.e.,

$$\left(\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times-\frac{\omega^2}{c^2}\right)\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)=\mathsf{1}\delta(\mathbf{r}-\mathbf{r}')\qquad(\mathbf{r}\in V), \tag{23}$$

from which it follows that

$$\frac{\omega^2}{c^2}\boldsymbol{\nabla}\cdot\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)=-\boldsymbol{\nabla}\delta(\mathbf{r}-\mathbf{r}')\qquad(\mathbf{r}\in V). \tag{24}$$

Hence from Eqs. (22) and (24) we derive

$$\begin{aligned}&\boldsymbol{\nabla}\cdot\mathrm{Im}\,\Delta\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)\\&\quad=-\boldsymbol{\nabla}\cdot\mathrm{Im}\,[\Delta\varepsilon(\mathbf{r},\omega)\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]\qquad(\mathbf{r}\in V),\end{aligned} \tag{25}$$

and thus from Eq. (19) the change $\Delta\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle$ is found to be

$$\begin{aligned}\Delta\langle\underline{\hat{\rho}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle&=-\frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\\&\times\,\boldsymbol{\nabla}\cdot\mathrm{Im}\,[\Delta\varepsilon(\mathbf{r},\omega)\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]\qquad(\mathbf{r}\in V).\end{aligned} \tag{26}$$

The change $\Delta\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle$ $(\mathbf{r}\in V)$ can be found in a similar way. Making use of Eqs. (22) and (23), from Eq. (20) we obtain

$$\begin{aligned}\Delta\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle&=-\frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\\&\times\,\mathrm{Im}\,[\Delta\varepsilon(\mathbf{r},\omega)\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]\times\overleftarrow{\boldsymbol{\nabla}}'\qquad(\mathbf{r}\in V),\end{aligned} \tag{27}$$

which implies that

$$\begin{aligned}&\Delta\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\times\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle\\&\quad=\frac{\hbar}{\pi}\frac{\omega^2}{c^2}\,\delta(\omega-\omega')\mathrm{Im}\,\{\Delta\varepsilon(\mathbf{r},\omega)\boldsymbol{\nabla}'\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]\\&\qquad-\Delta\varepsilon(\mathbf{r},\omega)\boldsymbol{\nabla}'\cdot\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)\}\qquad(\mathbf{r}\in V).\end{aligned} \tag{28}$$

Using Eqs. (26) and (28), we can now easily calculate, according to Eq. (12), the Casimir force acting on a dielectric body of volume V_{M} and dielectric susceptibility $\chi_{\mathrm{M}}(\mathbf{r},\omega)\equiv\Delta\varepsilon(\mathbf{r},\omega)$ (cf. Figs. 1 and 2) as

$$\begin{aligned}\mathbf{F}=&\frac{\hbar}{\pi c^2}\int_0^\infty \mathrm{d}\omega\,\omega^2\\&\times\,\mathrm{Im}\int_{V_{\mathrm{M}}}\mathrm{d}^3r\,\big\{\chi_{\mathrm{M}}(\mathbf{r},\omega)\boldsymbol{\nabla}'\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]\\&\quad-(\boldsymbol{\nabla}+\boldsymbol{\nabla}')\cdot\chi_{\mathrm{M}}(\mathbf{r},\omega)\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)\big\}_{\mathbf{r}'\to\mathbf{r}}.\end{aligned} \tag{29}$$

The reciprocity property of $\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)$ implies that $\mathrm{Tr}\,\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)$ is symmetric with respect to $\mathbf{r}$ and $\mathbf{r}'$. Thus we may rewrite Eq. (29) as

$$\begin{aligned}\mathbf{F}=&\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\omega\,\omega^2\\&\times\,\bigg\{\mathrm{Im}\int_{V_{\mathrm{M}}}\mathrm{d}^3r\,\chi_{\mathrm{M}}(\mathbf{r},\omega)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\\&\quad-2\,\mathrm{Im}\int_{\partial V_{\mathrm{M}}}\mathrm{d}\mathbf{a}\cdot\chi_{\mathrm{M}}(\mathbf{r},\omega)[\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\bigg\}.\end{aligned} \tag{30}$$

Further, on recalling the analytic properties of the integrands as functions of (complex) ω, we may employ contour integral techniques to represent Eq. (30) in the

form of

$$\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty d\xi\,\xi^2 \times \left\{\int_{V_\mathrm{M}} d^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} - 2\int_{\partial V_\mathrm{M}} d\mathbf{a}\cdot\chi_\mathrm{M}(\mathbf{r},i\xi)[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}\right\}. \quad (31)$$

In Eqs. (29)–(31) the coincidence limit $\mathbf{r}'\to\mathbf{r}$ has again to be performed in such a way that unphysical "self-force" contributions are removed.

If the body consists of homogeneous dielectric matter, one can simply replace the Green tensor $[\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}$ with its scattering part $\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega)$. In the case of inhomogeneous matter this replacement should be done point-wise. That is to say, at each space point $\mathbf{r}$, the Green tensor for the corresponding bulk material must be subtracted from $\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)$ in the limit $\mathbf{r}'\to\mathbf{r}$. In the case of an isolated body (cf. Fig. 1) the surface integral in Eqs. (30) and (31) can be dropped, hence

$$\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty d\xi\,\xi^2 \times \int_{V_\mathrm{M}} d^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}. \quad (32)$$

Clearly, the surface integral must not be dropped in the case where the body is an inner part of a larger dielectric body (cf. Fig. 2).

Equation (32) is the desired generalization of Eq. (4). In contrast to Eq. (4), it represents the exact force acting on a dielectric body of given permittivity, since $\chi_\mathrm{M}(\mathbf{r},i\xi)$ is not restricted to small values anymore. Correspondingly, the Green tensor in Eq. (32) is the one that takes the presence of the dielectric body fully into account, whereas in the Green tensor in Eq. (4) the presence of the dielectric body is not considered. Hence in contrast to Eq. (4), Eq. (32) includes in the calculation of the Casimir force that acts on a dielectric body the body's retroaction on the electromagnetic ground-state noise of the residual system.

To make this explicit, one can expand the full Green tensor $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in powers of $\Delta\varepsilon(\mathbf{r},i\xi)$ by using the iterative solution to Eq. (22) [together with Eq. (21)]. Inserting the resulting Born series for $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in Eq. (31), one obtains the corresponding expansion of the Casimir force $\mathbf{F}$ in powers of $\Delta\varepsilon(\mathbf{r},i\xi)$. In particular, truncating this expansion at the term linear in $\Delta\varepsilon(\mathbf{r},i\xi)$ [$\Delta\varepsilon(\mathbf{r},i\xi)\mapsto\delta\varepsilon(\mathbf{r},i\xi)$], i.e., replacing $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ with its zeroth-order approximation $\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)$, we simply obtain

$$\mathbf{F} = -\frac{\hbar}{2\pi c^2}\int_0^\infty d\xi\,\xi^2 \times \left\{\int_{V_\mathrm{M}} d^3r\,\chi_\mathrm{M}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} - 2\int_{\partial V_\mathrm{M}} d\mathbf{a}\cdot\chi_\mathrm{M}(\mathbf{r},i\xi)[\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}\right\}. \quad (33)$$

In this case, the prescription for taking the coincidence limit of the Green tensor simply consists in the replacement $[\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}\mapsto\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$, where $\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r}',i\xi)$ is the scattering part of the Green tensor $\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)$ in the absence of the dielectric matter in V (cf. Figs. 1 and 2). Note that if the surface integral can be dropped (i.e., if the case sketched in Fig. 1 is considered), then Eq. (33) becomes identical with Eq. (4). It should be mentioned that inclusion in Eq. (33) of the higher-order terms of the Born series of $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ generates an increasing number of many-body corrections.

It may be also informative to examine the effect of the change $\Delta\varepsilon(\mathbf{r},\omega)$ not only on the level of the expectation values in Eqs. (19) and (20), but more directly on the level of the corresponding operators. This is outlined in Appendix B for weakly dielectric material [$\Delta\varepsilon(\mathbf{r},\omega)\mapsto\delta\varepsilon(\mathbf{r},\omega)$]. In this context, an alternative derivation of Eq. (33) is given.

In view of Eq. (13), Eq. (31) can be also given in the form of a surface integral, where the stress tensor $\mathsf{T}(\mathbf{r})$ can be found by studying [in a similar way as in the derivation of Eq. (31)] the change induced in Eqs. (14) and (15) by the change $\Delta\varepsilon(\mathbf{r},i\xi)$. More easily, we may directly derive it (up to irrelevant transverse contributions) from Eq. (31) as

$$\mathsf{T}(\mathbf{r}) = \frac{\hbar}{\pi c^2}\int_0^\infty d\xi\,\xi^2\chi_\mathrm{M}(\mathbf{r},i\xi)[\mathsf{G}_V(\mathbf{r},\mathbf{r},i\xi)]_{\mathbf{r}'\to\mathbf{r}} + \frac{\hbar}{2\pi c^2}\boldsymbol{\nabla}\int_0^\infty d\xi\,\xi^2\int_{V_\mathrm{M}} d^3r' \times \frac{\chi_\mathrm{M}(\mathbf{r}',i\xi)}{4\pi\,|\mathbf{r}-\mathbf{r}'|}\boldsymbol{\nabla}'\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r}',\mathbf{r}'',i\xi)]_{\mathbf{r}''\to\mathbf{r}'}, \quad (34)$$

which makes obvious the fact that the stress tensor depends on the permittivity in a spatially non-local way in general. With respect to the volume integral in Eq. (34), $\mathbf{r}$ should be thought of as being infinitesimally outside V_M if necessary. The formulation of the force in terms of the stress tensor is particularly advantageous in the case of homogeneous material. From Eq. (31) it is easily seen that for a homogeneous body that is an inner part of a larger body (cf. Fig. 2) the somewhat cumbersome stress tensor (34) can be replaced with

$$\mathsf{T}(\mathbf{r}) = -\frac{\hbar}{\pi c^2}\int_0^\infty d\xi\,\xi^2\chi_\mathrm{M}(i\xi)\left\{\tfrac{1}{2}\mathsf{1}\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)] - \mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)\right\}_{\mathbf{r}'\to\mathbf{r}}, \quad (35)$$

and for an isolated homogeneous body (cf. Fig. 1) it follows from Eq. (32) that it can be replaced with

$$\mathsf{T}(\mathbf{r}) = -\frac{\hbar}{2\pi c^2}\int_0^\infty d\xi\,\xi^2\chi_\mathrm{M}(i\xi)\mathsf{1}\mathrm{Tr}\,[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}. \quad (36)$$

Furthermore, the assumed homogeneity then implies that we may let $[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}\mapsto\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ in Eqs. (35) and (36). Needless to say that replacing the full Green tensor $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ as appearing in Eqs. (34)–(36) with

the zeroth-order approximation $\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)$ yields again the Casimir force in the case of weakly dielectric material.

V. FORCE ON MICRO-OBJECTS AND ATOMS

Since nothing has been said about the spatial extension of the bodies under consideration, the applicability of Eqs. (31) and (32) ranges from dielectric macro-objects to micro-objects, even including single atoms. Let us consider a dielectric body that may be thought of as consisting of distinguishable (electrically neutral but polarizable) micro-constituents frequently called atoms or molecules within the framework of molecular optics. We may then assume the validity of the Clausius–Mosotti relation [23, 24],

$$\begin{aligned}\chi_{\mathrm{M}}(\mathbf{r},\omega) &= \varepsilon_0^{-1}\eta(\mathbf{r})\alpha(\omega)[1-\eta(\mathbf{r})\alpha(\omega)/(3\varepsilon_0)]^{-1}\\ &= \varepsilon_0^{-1}\eta(\mathbf{r})\alpha(\omega)\,[1+\chi_{\mathrm{M}}(\mathbf{r},\omega)/3],\end{aligned} \tag{37}$$

where $\alpha(\omega)$ is the polarizability of a single micro-constituent and $\eta(\mathbf{r})$ the number density of the micro-constituents (referred to as atoms in the following). It is worth noting that there is no need here—in contrast to Eq. (1)—to regard $\alpha(\omega)$ as being calculated in the lowest (non-vanishing) order of perturbation theory according to Eq. (2). It can be shown (Appendix C) that Eq. (37) is consistent with the requirement that both $\alpha(\omega)$ and $\chi_{\mathrm{M}}(\mathbf{r},\omega)$ be Fourier transforms of response functions iff

$$\eta(\mathbf{r})\alpha(0)/(3\varepsilon_0) < 1. \tag{38}$$

A. Isolated micro-object

Let V_{M} be the small volume of an isolated dielectric micro-object (cf. Fig. 1) which a dielectric susceptibility $\chi_{\mathrm{M}}(\omega)$ of Clausius–Mosotti-type can be ascribed to. Combining Eq. (32) with Eq. (37) and assuming that, due to the smallness of V_{M}, the scattering part of the Green tensor can be taken out of the space integral at the (appropriately chosen) position $\mathbf{r}$ of the micro-object, we derive the force acting on the micro-object to be

$$\begin{aligned}\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\left[1+\tfrac{1}{3}\chi_{\mathrm{M}}(i\xi)\right]\\ \times\,\boldsymbol{\nabla}\mathrm{Tr}\,\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi).\end{aligned} \tag{39}$$

Recall that in the case under study the replacement $[\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}} \mapsto \mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ can be made. Equation (39), which generalizes Eq. (6), differs in two respects from Eq. (6). Firstly, its validity is no longer restricted to weakly dielectric matter. Secondly, it takes into account the dependence of the force on the shape of the micro-object.

In contrast to Eq. (6), the force as given by Eq. (39) includes all-order multi-atom van der Waals interactions of the micro-object, as may be seen by expanding the Green tensor $\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi)$ in powers of $\chi_{\mathrm{M}}(i\xi)$ (cf. Ref. [13]). If they are disregarded, Eq. (39) reduces to (Appendix D)

$$\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\boldsymbol{\nabla}\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi), \tag{40}$$

which, as expected, is nothing but Eq. (6)—only the term linear in $\alpha(i\xi)$ contributes to the force. The force in this limit is simply the sum of the forces acting on the atoms due to the presence of the external bodies (region V_{B} in Fig. 1). Hence, $\mathbf{F}^{(\mathrm{at})} = (V_{\mathrm{M}}\eta)^{-1}\mathbf{F}$ is the force acting on a single ground-state atom, that is to say, we are left exactly with the formula for the CP force as given by Eq. (1), with the exception that now the atomic polarizability is the exact one rather than the perturbative expression given in Eq. (3).

B. Micro-object that is an inner part of a larger body

Let now V_{M} be the small volume of a dielectric micro-object that belongs to a larger body of volume V of the same atoms (cf. Fig. 2). Under assumptions analogous to those leading from Eq. (32) to Eq. (39), from Eq. (31) [together with Eq. (37)] we obtain the following formula for the (shape-dependent) force acting on the micro-object:

$$\begin{aligned}\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\left[1+\tfrac{1}{3}\chi_{\mathrm{M}}(i\xi)\right]\\ \times\,\boldsymbol{\nabla}\cdot\left[\tfrac{1}{2}\mathsf{1}\mathrm{Tr}\,\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)-\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)\right].\end{aligned} \tag{41}$$

Equation (41) differs from Eq. (39) in the second term in the square brackets in the second line. This difference can be regarded as reflecting the fact that—in contrast to Eq. (39)—the force acting on the micro-object is screened by the residual part of the body.

When the multi-atom van der Waals interactions of the body (of volume V) are disregarded, then, in a way quite similar to that outlined in the derivation of Eq. (40) in App. D, Eq. (41) can be shown to reduce to the term linear in the atomic polarizability,

$$\begin{aligned}\mathbf{F} = -V_{\mathrm{M}}\eta\,\frac{\hbar\mu_0}{\pi}\int_0^\infty \mathrm{d}\xi\,\xi^2\,\alpha(i\xi)\\ \times\,\boldsymbol{\nabla}\cdot\left[\tfrac{1}{2}\mathsf{1}\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)-\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)\right].\end{aligned} \tag{42}$$

Recall that in this approximation $\mathsf{G}_V^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$ can be replaced with $\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$. From Eq. (42) it then follows that $\mathbf{F}^{(\mathrm{at})} = (V_{\mathrm{M}}\eta)^{-1}\mathbf{F}$ can be regarded as the screened CP force acting on an atom of a weakly dielectric medium.

To make contact with earlier results, let us apply Eq. (42) to the atoms of a weakly dielectric medium (corresponding to the region V in Fig. 2) in front of a laterally infinitely extended magnetodielectric planar wall (corresponding to the region V_{B} in Fig. 2), which is assumed to

extend from some negative z value up to $z=0$. Using the explicit form of the Green tensor for planar multi-layer structures (see, e.g., [25, 26]), we may write its scattering part for coincident spatial arguments in the (empty) space region $z>0$ as

$$\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega)=\frac{i}{8\pi^2k^2}\int \mathrm{d}^2q\,\frac{e^{2i\beta z}}{\beta}\left\{r_-^p\left[q^2\mathbf{e}_z\mathbf{e}_z - \beta^2\mathbf{e}_q\mathbf{e}_q\right]+r_-^s k^2\mathbf{e}_s\mathbf{e}_s\right\}, \quad (43)$$

with $k=k(\omega)=\omega/c$, $q=|\mathbf{q}|$, $\beta=\beta(\omega,q)=(k^2-q^2)^{1/2}$ and orthogonal unit vectors $\mathbf{e}_q=\mathbf{q}/q$, $\mathbf{e}_z=\boldsymbol{\nabla}z$, and $\mathbf{e}_s=\mathbf{e}_q\times\mathbf{e}_z$. The effect of the (multi-layered) wall is described in terms of the generalized reflection coefficients $r_-^\sigma=r_-^\sigma(\omega,q)$ $(\sigma=s,p)$, which in the simplest case of an internally homogeneous, semi-infinite wall reduce to the usual Fresnel amplitudes. From Eq. (43) it then follows that

$$\mathrm{Tr}\,\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},\omega)=\frac{i}{4\pi}\int_0^\infty \mathrm{d}q\,q\,\frac{e^{2i\beta z}}{\beta}\left[(r_-^s-r_-^p)+\frac{2q^2}{k^2}\,r_-^p\right]. \quad (44)$$

Substitution of Eq. (44) into Eq. (40) leads to the well-known expression [14, 15, 16, 17, 18] for the CP force acting on a single ground-state atom in front of a planar wall. The screened force acting on a weakly dielectric medium atom is obtained by substituting Eqs. (43) and (44) into Eq. (42). The result is $(\beta=i\kappa)$

$$\begin{aligned}\mathbf{F}^{(\mathrm{at})}(z)&=(V_\mathrm{M}\eta)^{-1}\mathbf{F}(z)\\ &=\mathbf{e}_z\frac{\hbar\mu_0}{4\pi^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\alpha(i\xi)\int_0^\infty \mathrm{d}q\,qe^{-2\kappa z}(r_-^s-r_-^p). \quad (45)\end{aligned}$$

It fully agrees with the result found by calculating the Casimir stress (14) [together with Eq. (15)] in a dielectric layer of a planar multi-layer structure and performing therein the limit to weakly dielectric matter [4, 5].

VI. VAN DER WAALS INTERACTION BETWEEN TWO ATOMS

Equation (31) can also be regarded as a basic equation for calculating the force between two (ground-state) atoms. For this purpose, let us consider the small change $\delta\mathbf{F}$ of $\mathbf{F}$ in Eq. (31) due to a small change $\delta\chi_1(\mathbf{r},i\xi)$ of the susceptibility $\chi_\mathrm{M}(\mathbf{r},i\xi)$ and a small change $\delta\chi_2(\mathbf{r},i\xi)$ of the susceptibility $\chi_\mathrm{B}(\mathbf{r},i\xi)$ (of one of the bodies) in the region V_B in Fig. (2). In particular let us assume that $\chi_\mathrm{M}(\mathbf{r},i\xi)$ only changes inside the region V_M. Recalling Eq. (22), it is not difficult to calculate $\delta\mathbf{F}$ up to second order in $\delta\chi_k(\mathbf{r},i\xi)$ $(k=1,2)$ and pick out the term $\delta_{12}\mathbf{F}$ that is bilinear in $\delta\chi_1(\mathbf{r},i\xi)$ and $\delta\chi_2(\mathbf{r},i\xi)$:

$$\begin{aligned}\delta_{12}\mathbf{F}&=\frac{\hbar}{2\pi c^4}\int_0^\infty \mathrm{d}\xi\,\xi^4\int_{V_\mathrm{M}}\mathrm{d}^3r\,\delta\chi_1(\mathbf{r},i\xi)\\ &\times\int_{V_\mathrm{B}}\mathrm{d}^3s\,\delta\chi_2(\mathbf{s},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\left[\mathsf{G}_V(\mathbf{r},\mathbf{s},i\xi)\cdot\mathsf{G}_V(\mathbf{s},\mathbf{r},i\xi)\right], \quad (46)\end{aligned}$$

where the Green tensor $\mathsf{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)$ refers to the system before the susceptibilities have been changed. Note that, since we are dealing with the interaction between two well-separated space regions, the problem of removing "self"-force contributions does not arise here.

Now let us suppose that the small changes $\delta\chi_1(\mathbf{r},i\xi)$ and $\delta\chi_2(\mathbf{r},i\xi)$ result from the introduction into the system of additional atoms, say impurity atoms, of type 1 and type 2, respectively. The (body-assisted) force acting on a type-1 atom at position $\mathbf{r}_1$ due to its interaction with a type-2 atom at position $\mathbf{r}_2$ is then evidently obtained, in first order of their polarizabilities $\alpha_1(i\xi)$ and $\alpha_2(i\xi)$, from the "crossing term" $\delta_{12}\mathbf{F}$ as

$$\begin{aligned}\mathbf{F}_{12}^{(\mathrm{at})}&=\frac{\hbar\mu_0^2}{2\pi}\int_0^\infty \mathrm{d}\xi\,\xi^4\alpha_1(i\xi)\alpha_2(i\xi)\\ &\times\boldsymbol{\nabla}_1\mathrm{Tr}\left[\mathsf{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)\cdot\mathsf{G}_V(\mathbf{r}_2,\mathbf{r}_1,i\xi)\right], \quad (47)\end{aligned}$$

which is in full agreement with previous calculations of the van der Waals interaction between two atoms [27, 28, 29]. Recall that $\mathsf{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)$ is the Green tensor for the material system that was present before the introduction of the additional atoms.

Disregarding local-field corrections, one may insert in Eq. (47) the Green tensor for the unperturbed host media. In particular, the force between two atoms embedded in a homogeneous (dielectric) background medium is then obtained by identifying $\mathsf{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)$ with the well-known bulk-medium Green tensor. Note that in this case the same formula for the force can be obtained by basing the calculations on the Minkowski stress tensor [5, 11, 30]. Choosing in Eq. (47) the free-space Green tensor, we recover the van der Waals interaction between two atoms in otherwise empty space. It should be pointed out that $\mathbf{F}_{12}^{(\mathrm{at})}$ and $\mathbf{F}_{21}^{(\mathrm{at})}$ obey the lex tertia $\mathbf{F}_{12}^{(\mathrm{at})}=-\mathbf{F}_{21}^{(\mathrm{at})}$ if the Green tensor is translationally invariant $[\mathsf{G}_V(\mathbf{r}_1+\mathbf{v},\mathbf{r}_2+\mathbf{v},i\xi)=\mathsf{G}_V(\mathbf{r}_1,\mathbf{r}_2,i\xi)]$, as it is the case for the two atoms being in bulk material or in free space. Since Eq. (47) describes the atom–atom force in the presence of arbitrary macroscopic bodies, it is clear that the atomic positions $\mathbf{r}_1$ and $\mathbf{r}_2$ are not physically equivalent in general.

VII. SUMMARY AND CONCLUSIONS

Within the framework of macroscopic QED in linearly, locally, and causally responding media, we have shown that dispersive forces acting on (ground-state) macro- and micro-objects—including single atoms—can be calculated in a unified way on the basis of the Lorentz force density that acts on the charge and current densities attributed to the polarization and magnetization of the media. Although the examples considered in Secs. IV–VI refer to dielectric objects, the basic formulas given in Sec. III can also be used to include in the calculations magnetic properties of the matter. Inclusion in the theory of non-locally responding media would require an

extension of the underlying quantization scheme, which may be a subject of further studies.

We have derived very general formulas for the force acting on a dielectric body or a part of it—formulas which apply to arbitrary geometries and whose validity is not restricted to weakly dielectric matter. For locally responding dielectric matter that may be regarded as consisting of atoms in the broadest sense of the word, the permittivity can be assumed to be of Clausius–Mosotti-type. In this way, all relevant many-atom van der Waals interactions of the involved matter can be included in the force to be calculated.

As already mentioned, the applicability of the theory ranges from macro-objects to micro-objects. Commonly, the force acting on a (weakly) dielectric micro-object is calculated in the spirit of a simple superposition of CP forces acting on independent atoms. The present theory enables one to systematically include in the calculation both the dependence of the force on the shape of the micro-object and, at the same time, the contributions to the force due to many-atom interactions of atoms of the micro-object, without restriction to weakly dielectric matter.

If the micro-object reduces to a single atom, the well-known formula for the CP force on a single atom is recovered. It is worth noting that not only the force acting on an isolated atom can be obtained, but also the force on a medium atom. For a medium atom, the CP force is screened due to the presence of neighboring medium atoms, while there is of course no such screening in the case of an isolated atom. Moreover, the basic formulas can also be used to study the body-assisted van der Waals interaction between atoms.

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APPENDIX A: DERIVATION OF EQS. (16)-(18)

To express $\underline{\hat{\rho}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)$ as defined by Eqs. (8) and (9), respectively, in terms of $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$, we first insert Eqs. (10) and (11) in Eqs. (8) and (9). Taking into account that the Green tensor obeys the differential equation

$$\boldsymbol{\nabla}\times\kappa(\mathbf{r},\omega)\boldsymbol{\nabla}\times\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)-\frac{\omega^2}{c^2}\varepsilon(\mathbf{r},\omega)\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)=\mathbf{1}\delta(\mathbf{r}-\mathbf{r}') \tag{A1}$$

(together with the boundary condition at infinity) as well as the relation that follows by taking the divergence of Eq. (A1),

$$\frac{\omega^2}{c^2}\,\boldsymbol{\nabla}\cdot[\varepsilon(\mathbf{r},\omega)\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)]=-\boldsymbol{\nabla}\delta(\mathbf{r}-\mathbf{r}'), \tag{A2}$$

we easily see by straightforward calculation that Eqs. (16) and (17) hold. According to Ref. [19], the noise current expressed in terms of the noise polarization and the noise magnetization,

$$\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)=-i\omega\underline{\hat{\mathbf{P}}}_{\mathrm{N}}(\mathbf{r},\omega)+\boldsymbol{\nabla}\times\underline{\hat{\mathbf{M}}}_{\mathrm{N}}(\mathbf{r},\omega), \tag{A3}$$

can be related to bosonic fields $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)$ ($\lambda=e,m$),

$$\left[\hat{f}_{\lambda k}(\mathbf{r},\omega),\hat{f}^\dagger_{\lambda' l}(\mathbf{r}',\omega')\right]=\delta_{kl}\delta_{\lambda\lambda'}\delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'), \tag{A4}$$

by means of the relations

$$\underline{\hat{\mathbf{P}}}_{\mathrm{N}}(\mathbf{r},\omega)=i\left[\hbar\varepsilon_0\mathrm{Im}\,\varepsilon(\mathbf{r},\omega)/\pi\right]^{1/2}\hat{\mathbf{f}}_e(\mathbf{r},\omega), \tag{A5}$$

$$\underline{\hat{\mathbf{M}}}_{\mathrm{N}}(\mathbf{r},\omega)=\left[-\hbar\kappa_0\mathrm{Im}\,\kappa(\mathbf{r},\omega)/\pi\right]^{1/2}\hat{\mathbf{f}}_m(\mathbf{r},\omega). \tag{A6}$$

Note that the $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger_\lambda(\mathbf{r},\omega)$ play the role of the basic variables of the combined system composed of the electromagnetic field and the (linear) medium. Inserting Eqs. (A5) and (A6) in Eq. (A3) and making use of Eq. (A4) we find that $\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}^\dagger_{\mathrm{N}}(\mathbf{r},\omega)$ obey the commutation relation

$$\begin{aligned}\left[\underline{\hat{j}}_{\mathrm{N}k}(\mathbf{r},\omega),\underline{\hat{j}}^\dagger_{\mathrm{N}l}(\mathbf{r}',\omega')\right]&=\frac{\hbar}{\mu_0\pi}\,\delta(\omega-\omega')\\&\times\Bigg[\frac{\omega^2}{c^2}\sqrt{\mathrm{Im}\,\varepsilon(\mathbf{r},\omega)}\,\mathbf{1}\delta(\mathbf{r}-\mathbf{r}')\sqrt{\mathrm{Im}\,\varepsilon(\mathbf{r}',\omega')}\\&+\boldsymbol{\nabla}\times\sqrt{\mathrm{Im}\,\kappa(\mathbf{r},\omega)}\,\mathbf{1}\delta(\mathbf{r}-\mathbf{r}')\sqrt{\mathrm{Im}\,\kappa(\mathbf{r}',\omega')}\times\overleftarrow{\boldsymbol{\nabla}}'\Bigg]_{kl},\end{aligned} \tag{A7}$$

which immediately implies the expression for the ground-state expectation value $\langle\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}^\dagger_{\mathrm{N}}(\mathbf{r}',\omega')\rangle$ as given in Eq. (18).

APPENDIX B: ALTERNATIVE DERIVATION OF EQ. (33)

Let us consider a small variation $\delta\varepsilon(\mathbf{r},\omega)$ and the corresponding (first-order) changes of the operators $\underline{\hat{\rho}}(\mathbf{r},\omega)$, $\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)$, $\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)$, and $\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega)$, and assume that $\delta\varepsilon(\mathbf{r},\omega)$ is different from zero only in the volume V [cf. Fig. 2]. From Eqs. (8)-(11) it follows that

$$\begin{aligned}\delta\underline{\hat{\rho}}(\mathbf{r},\omega)=&-\varepsilon_0\boldsymbol{\nabla}\cdot\{\delta\varepsilon(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\\&+[\varepsilon(\mathbf{r},\omega)-1]\delta\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\}+(i\omega)^{-1}\boldsymbol{\nabla}\cdot\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega),\end{aligned} \tag{B1}$$

$$\begin{aligned}\delta\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)=&-i\omega\varepsilon_0\{\delta\varepsilon(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\\&+[\varepsilon(\mathbf{r},\omega)-1]\delta\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\}+\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega),\end{aligned} \tag{B2}$$

$$\delta\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) = i\mu_0\omega \int \mathrm{d}^3 s \left[\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega) + \delta\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega)\right], \quad \text{(B3)}$$

$$\delta\underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = \mu_0 \boldsymbol{\nabla}\times \int \mathrm{d}^3 s \left[\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega) + \delta\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{s},\omega)\right]. \quad \text{(B4)}$$

Since the state space attributed to the dynamical variables $\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)$, in terms of which the electromagnetic quantities are thought of as being expressed, can be regarded as being independent of the chosen permittivity (and/or permeability) [19], we may apply the rule $\langle\delta\cdots\rangle = \delta\langle\cdots\rangle$ when calculating expectation values. Thus, combining Eqs. (8)-(11) with Eqs. (B1)-(B4), we derive

$$\begin{aligned}\delta\langle\rho(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle &= -\varepsilon_0\boldsymbol{\nabla}\cdot[\delta\varepsilon(\mathbf{r},\omega)\langle\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle] \\ &+ \frac{1}{i\omega}\boldsymbol{\nabla}\cdot[\langle\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle + \langle\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\delta\underline{\hat{\mathbf{E}}}^\dagger(\mathbf{r}',\omega')\rangle] \\ &+ \dots \end{aligned} \quad \text{(B5)}$$

and

$$\begin{aligned}\delta\langle\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle &= -i\omega\varepsilon_0\delta\varepsilon(\mathbf{r},\omega)\langle\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle \\ &+ [\langle\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle + \langle\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\delta\underline{\hat{\mathbf{B}}}^\dagger(\mathbf{r}',\omega')\rangle] \\ &+ \dots, \end{aligned} \quad \text{(B6)}$$

where terms that vanish when $\mathbf{r},\mathbf{r}'\in V$ have not been quoted [$\varepsilon(\mathbf{r},\omega)=1$ in V]. Now from Eq. (12) together with Eqs. (B5) and (B6) and Eqs. (10), (11), (18), (B3), and (B4) we may easily calculate $\delta\mathbf{F}$. On recalling standard properties of the Green tensor, we derive

$$\delta\mathbf{F} = \delta^{(1)}\mathbf{F} + \delta^{(2)}\mathbf{F}, \quad \text{(B7)}$$

where

$$\begin{aligned}\delta^{(1)}\mathbf{F} &= \frac{\hbar}{2\pi}\int_0^\infty \mathrm{d}\omega\,\frac{\omega^2}{c^2} \\ &\times\left\{\int_{V_{\mathrm{M}}}\mathrm{d}^3 r\,\delta\varepsilon(\mathbf{r},\omega)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathrm{Im}\,\mathsf{G}(\mathbf{r},',\omega)]_{\mathbf{r}'\to\mathbf{r}} \right. \\ &\left. -2\int_{\partial V_{\mathrm{M}}}\mathrm{d}\mathbf{a}\cdot\delta\varepsilon(\mathbf{r},\omega)\mathrm{Im}\,[\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\right\} \end{aligned} \quad \text{(B8)}$$

arises from the first terms on the right-hand sides of Eqs. (B5) and (B6), and

$$\begin{aligned}\delta^{(2)}\mathbf{F} &= \frac{\hbar}{2\pi}\int_0^\infty \mathrm{d}\omega\,\frac{\omega^2}{c^2} \\ &\times\left\{\int_{V_{\mathrm{M}}}\mathrm{d}^3 r\,[\mathrm{Im}\,\delta\varepsilon(\mathbf{r},\omega)]\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}^*(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}} \right. \\ &\left. -2\int_{\partial V_{\mathrm{M}}}\mathrm{d}\mathbf{a}\cdot[\mathrm{Im}\,\delta\varepsilon(\mathbf{r},\omega)][\mathsf{G}^*(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\right\} \end{aligned} \quad \text{(B9)}$$

arises from the second terms on the right-hand sides of Eqs. (B5) and (B6). Note that in the derivation of Eq. (B9), the relation

$$\begin{aligned}\langle\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\underline{\hat{\mathbf{j}}}_{\mathrm{N}}^\dagger(\mathbf{r}',\omega')\rangle &= \langle\underline{\hat{\mathbf{j}}}_{\mathrm{N}}(\mathbf{r},\omega)\delta\underline{\hat{\mathbf{j}}}_{\mathrm{N}}^\dagger(\mathbf{r}',\omega')\rangle \\ &= \frac{\hbar}{2\mu_0\pi}\delta(\omega-\omega')\frac{\omega^2}{c^2}\,\mathrm{Im}\,\delta\varepsilon(\mathbf{r},\omega)\mathsf{1}\delta(\mathbf{r}-\mathbf{r}') \end{aligned} \quad \text{(B10)}$$

has been used, which follows from Eq. (18). Combining Eqs. (B7), (B8), and (B9) yields

$$\begin{aligned}\delta\mathbf{F} &= \frac{\hbar}{2\pi}\int_0^\infty \mathrm{d}\omega\,\frac{\omega^2}{c^2} \\ &\times\,\mathrm{Im}\left\{\int_{V_{\mathrm{M}}}\mathrm{d}^3 r\,\delta\varepsilon(\mathbf{r},\omega)\boldsymbol{\nabla}\mathrm{Tr}\,[\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}} \right. \\ &\left. -2\int_{\partial V_{\mathrm{M}}}\mathrm{d}\mathbf{a}\cdot\delta\varepsilon(\mathbf{r},\omega)[\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)]_{\mathbf{r}'\to\mathbf{r}}\right\}. \end{aligned} \quad \text{(B11)}$$

Changing in Eq. (B11) the real-frequency integral to an imaginary-frequency integral in the usual way, we just arrive at Eq. (33) [$\delta\varepsilon(\mathbf{r},i\xi)\mapsto\chi_{\mathrm{M}}(\mathbf{r},i\xi)$, $\delta\mathbf{F}\mapsto\mathbf{F}$]. Note that only the real parts of Eqs. (B8) and (B9) contribute to Eq. (B11), while the imaginary parts drop out.

APPENDIX C: CLAUSIUS–MOSOTTI SUSCEPTIBILITY AND CAUSALITY

Since $\alpha(\omega)$ is the Fourier transform of a response function, it is holomorphic and without zeros in the upper complex half-plane (see, e.g., Ref. [20] for a summary of response function properties). Consequently, $\chi_{\mathrm{M}}(\mathbf{r},\omega)$ as given by (the first line of) Eq. (37) is there also holomorphic and without zeros, except for possible poles at ω-values satisfying the equation $\eta(\mathbf{r})\alpha(\omega)/(3\varepsilon_0)=1$. However, since $\alpha(\omega)$ is the Fourier transform of a response function, it is real only on the imaginary frequency axis, where it, beginning with a positive value at $\omega=0$, monotonically decreases with increasing imaginary frequency. Hence, if $\eta(\mathbf{r})\alpha(0)/(3\varepsilon_0)\geq 1$ is valid, a pole is observed and $\chi_{\mathrm{M}}(\mathbf{r},\omega)$ would fail to be a response function. From the requirement that both $\alpha(\omega)$ and $\chi_{\mathrm{M}}(\mathbf{r},\omega)$ be Fourier transforms of response functions, it thus follows that the condition $\eta(\mathbf{r})\alpha(0)/(3\varepsilon_0)<1$ must be imposed on Eq. (37).

APPENDIX D: DERIVATION OF EQ. (40)

In order to derive Eq. (40), we return to Eq. (32) together with Eq. (37) and recall that, according to Eqs. (21) and (22), the Green tensor $\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega)$ obeys the equation

$$\begin{aligned}\mathsf{G}_V(\mathbf{r},\mathbf{r}',\omega) &= \mathsf{G}(\mathbf{r},\mathbf{r}',\omega) \\ &+ \frac{\omega^2}{c^2}\int_{V_{\mathrm{M}}}\mathrm{d}^3 s\,\mathsf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\chi_{\mathrm{M}}(\mathbf{s},\omega)\mathsf{G}_V(\mathbf{s},\mathbf{r}',\omega). \end{aligned} \quad \text{(D1)}$$

According to the principal volume method [31], the decomposition

$$\mathsf{G}(\mathbf{r},\mathbf{s},\omega) = \mathcal{P}\mathsf{G}(\mathbf{r},\mathbf{s},\omega) - \frac{c^2}{\omega^2}\,\mathsf{L}\delta(\mathbf{r}-\mathbf{s}) \tag{D2}$$

is applicable to the Green tensor $\mathsf{G}(\mathbf{r},\mathbf{s},\omega)$ in Eq. (D1). The procedure to be followed is to choose first an exclusion volume whose shape determines the tensor L, and to subsequently treat integrals over $\mathcal{P}\mathsf{G}(\mathbf{r},\mathbf{r}',\omega)$ as (shape-dependent) principal value integrals over an exclusion volume with the specified shape (see, e.g., Ref. [26] for details). Using Eq. (D2) in Eq. (D1), and adopting a spherical exclusion volume for which $\mathsf{L}=1/3$, we have

$$\begin{aligned}\mathsf{G}_V(\mathbf{r},\mathbf{r}',i\xi) &= [1+\chi_{\mathrm{M}}(\mathbf{r},i\xi)/3]^{-1}\big[\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)\\ &-\frac{\xi^2}{c^2}\int_{V_{\mathrm{M}}}\mathrm{d}^3s\,\mathcal{P}\mathsf{G}(\mathbf{r},\mathbf{s},i\xi)\cdot\chi_{\mathrm{M}}(\mathbf{s},i\xi)\mathsf{G}_V(\mathbf{s},\mathbf{r}',i\xi)\big].\end{aligned} \tag{D3}$$

When inserted in Eq. (32), the second term on the right-hand side of Eq. (D3) leads to a double integral over $\mathbf{r}$ and $\mathbf{s}$. Since contributions with $\mathbf{s}=\mathbf{r}$ are left out in the principal value integral, the corresponding part of the force is associated with at least two-atom interactions in V_{M}. Dropping all these multi-atom contributions, we obtain

$$\begin{aligned}\mathbf{F} = &-\frac{\hbar}{2\pi c^2}\int_0^\infty \mathrm{d}\xi\,\xi^2\\ &\times\int_{V_{\mathrm{M}}}\mathrm{d}^3r\,\chi_{\mathrm{M}}(\mathbf{r},i\xi)\boldsymbol{\nabla}\mathrm{Tr}\left[\frac{\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)}{1+\chi_{\mathrm{M}}(\mathbf{r},i\xi)/3}\right]_{\mathbf{r}'\to\mathbf{r}}.\end{aligned} \tag{D4}$$

Applying Eq. (D4) to a micro-object whose number density of atoms is constant over the small volume V_{M}, so that a position-independent Clausius-Mosotti susceptibility according to Eq. (37) can be assigned to it, and replacing $[\mathsf{G}(\mathbf{r},\mathbf{r}',i\xi)]_{\mathbf{r}'\to\mathbf{r}}$ with $\mathsf{G}^{(\mathrm{S})}(\mathbf{r},\mathbf{r},i\xi)$, we just arrive at Eq. (40).

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